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Thinking towards synergistic green refineries

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Abstract

The switch from a mined-carbon-based society to a bio-carbon based society requires a major shift not only of the resources, but also in the type of processes that produce the products on which our civilization builds; a richer approach to handling and production is required. Natural products from renewable sources will substitute today's purely synthetic products from fossil resources, associated waste streams will have to be utilized for the production of usable materials, with chemical processing being one of the main options.

Fuel represents a main class of chemicals that we increasingly rely on. They have to be substituted as quick as possible as they represent the largest use of mined carbon. The paper presents some of the stumbling blocks which prohibit the transfer and high lights the most needed research objectives.

Keywords: Biorefinery, Green refinery, Biobased Economy

1. Status and proposition

On at least part of humanity the light has dawned that fossil carbon sources are limited and that the mined carbon is currently largely dispatched into the atmosphere. Also, quite obviously the life cycle of the mined carbon is not closed and the biosphere has not adjusted to the increased load of carbon dioxide. In addition, rather then enabling the biosphere to increase its capacity to bind carbon dioxide; humans are busy to reduce its CO_2-storage capacity in the attempt to increase food production and harvesting trees for economical reasons. Whilst the carbon-dioxide problematic is known since at least the 1980ties [1], it took more than 20 years for the public to accept the effect CO_2 emissions have on the climate. The issues around the composition of the atmosphere being observed these days are in an order of magnitude more severe than for example the nitrous oxides (NO_x) and chlorofluorcarbons (CFC's) problems detected in the 1960's and 70's. This simply because most of the large energy resources are attached to the C-household making the atmosphere-related problems much more severe and consequently more difficult to handle.

Nature has a large resilience to environmental disturbances and life on earth has proven to be long-term stable, but in different configurations. Changing to another relatively stationary situation may be catastrophic

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for large parts of the existing biosphere; including the human species. Further, what we consider small changes in the conditions may prove to trigger large-scale changes that consequently could lead to another stable state. The knowledge, gained over the past few decades must motivate us to operate within more conservative boundaries. A more sustainable behaviour should stabilize the current balance state and should inhibit triggering a change that we have no control over.

The human "success" is based on an aggressive growth, which in the situation of limited resources must be harnessed or it will lead to conflicts. The issue of available resources cannot be solved without a globe-scale compromise, which imposes constraints on how the human race is interacting with its environment. This stage has become global in such a short period of time that adaptation to a non-aggressive mode of operation is hard to achieve. Regulatory measures are required and cannot be left to a competing economy-driven society.

Greed is one of the main obstacles to overcome, which some social layers are starting to recognise as one of the main source of problems in effectively controlling our future. Consequently this behaviour pattern is recently being addressed more seriously [2]. Politics and economics are very tightly interlaced and with both being greedy in their own way, but mutually benefiting, it requires a change that is hard to achieve. Politics needs to impose the necessary regulatory and political changes to get control over the development that avoids catastrophic behaviour patterns. Regulatory forces are necessary to ensure that the system changes. Such forces are felt by the public and may not necessarily result in changes that are popular locally in a political/social context.

Some industries have felt this regulatory pressure (i.e. permit to operate) much earlier than others and have had to adjust accordingly. For example, the US paper giant Weyerhaeuser declared in the last century the "ecologically invisible plant", thereby suggesting a closure of their operations within the part of the biosphere it is operating in. This included the growth of trees, the manufacturing process in terms of plant waste output such as liquid-pipe waste, being mostly water, and stack-waste including obnoxious gases and small particles.

We suggest to adopt two largely technique-motivated objectives:

- *Consumption must meet the production in the same time scale, both in material and energy.*
- *Production must be optimized with the earth as the ultimate system.*

Further, the political bodies need to be aligned so as to drive the industrial developments towards these objectives. Whilst the achievement of these objectives cannot be ultimate, a move towards a sustainable state could reduce the danger of a catastrophic sequence of events, and provide the necessary time to learn more about how we can get better control over the effects of the human presence in an environment with limited resources.

2. Integrating into the biosphere

With an increasing awareness, the public will have to move towards a substitution of the main resources, namely the replacement of fossil fuel with bio-fuel and petrochemistry by biochemistry.

2.1. Fuel

Fuels are the single largest fossil-based products and their replacement would consequently have the largest impact on CO_2 emissions from mined carbon. The biosphere has been the main source of fuel before and in the beginning of industrialization. Wood in particular was a major source. It is only after the first world war that petrol became the main fuel source, mainly driven by its ease of production and handling.

Most common fuels are relatively small, simple molecules that can be oxidized releasing a significant amount of energy as part of the conversion. Products are simple oxides, like water in case of oxidizing hydrogen or carbon monoxide/dioxide in case of carbon. Production of fuel is thus focusing on small molecules releasing a high energy in its conversion process. Such molecules are small hydrocarbons and their derivatives and hydrogen.

The formation of oil is a slow process modifying the complex chemical compounds of the accumulated biomass evolving from the biosphere. Synthetic fuel production reproduces some of these processes, though not as a copy, but substitutes that lead to similar results with intermediates that are for example sugars of various kinds. Such biochemical conversion processes digest cellulose and hemicellulose at elevated temperature in presence of among other alternatives strong acids into glucose. This process has been known for a long time [3]. Once the sugar is available, several alternative routes are possible to generate fuels, one of the more common ones is the digestion to alcohols, mainly ethanol. An alternative synthesis pathways is thermochemical conversion, generally involving pyrolysis and/or gasification to achieve building blocks (generally CO and H_2) which subsequently are recombined catalytically to higher alkanes by e.g. Fischer Tropsch Synthesis [4].

With the stakes being high, activities in this field have grown rapidly in the last decade. Many of the developments are by no means new, as the suggestion of wood-based and bio-waste-based fuel production has periodically been funded, last in the 1970ties/80ties. In each period, new results have been produced, due to changes in technology and improved understanding as well as simply exploring a larger set of alternatives.

2.2. The stumbling block

If in fact the technology is available, why is it not used? The answer is quite simple and has been expressed very explicitly by W. Banholzer in his recent AIChE plenary lecture [5]: *economy*. The prices for fuel derived from petrol are much lower than the fuel production costs from biomass. For the petrol-based industry planning periods are generally weeks and months, very long-term planning has a horizon of 10 years. Stakeholders request pay-back times of years, thus the economical horizon is simply too short to accommodate the developments required to move towards a bio-based economy. The investments are simply too small to make the necessary impact at this point in time. Banholzer is a typical representative of the current leadership in the chemical industry who follows the rules of economy aiming at maximizing the profit for the company and, in the worst case, maintain the growth of the company, which is a necessary condition for survival [6, 7, 8]: *"The innovation pipeline must maintain a constant and predictable profit flow for a company to remain successful, particularly during turbulent economic times."*

Who is to blame? Banholzer clearly exhibits the horizons in which industry is operating [7]: economic cycles are 6-8 years and product development in the order of 5-20 years. The development of new technologies with an alternative feedstock AND a change of the process will take more likely in the order of 50 years. It is thus mainly the economical system that prohibits the developments over a longer time horizon. Simply exploring the domain is also not supported as industrial and private funding is black/white: Projects that potentially have a payback in the 10 years or shorter time horizon are being supported, whilst the others are discontinued. Additionally, public research funding increasingly follows the same essential guidelines, thus work is progressing at a very low flame.

In the current situation it is thus apparent that bio-fuels are economically not viable and only political pressure and long term incentives will possibly yield a move. However, if fuel production is combined with the production of chemicals, the economical aspect will change for the better.

2.3. Chemicals

The chemical industry started largely off using wood as a main resource [9, 10, 11]. In 1949, Glesinger published his book *"The coming age of wood"* describing an economy that makes use of wood in the large scale [12]. The timing for the book was unfortunate, as the petrol-based industry was just about to go into overdrive. Wood chemistry has been well-developed in the early 1900, but has nearly gone lost with the advent of petrol-based chemistry.

The large-scale industrialisation builds on petro-chemical products and a change to another supply will require major changes. Current developments mainly aim at replacing the intermediates and to continue feeding into the existing down-stream processes thereby reducing the risk and the development time. However, this approach is largely based on a destructive approach in the sense of breaking down the complex biomolecules into the small constituents from which the intermediates are being synthesized. It should be obvious to any science educated person that this leads to a waste of energy stored in form of complexity in the molecules. Statements as made by Banholzer that *"Biological systems reduce oxidized, lower-enthalpy carbon atoms in carbohydrates to ethanol by further oxidizing 33 % of the carbon atoms to CO_2. The simultaneous oxidation and reduction inherent in fermentation results in an overall poor carbon efficiency."* are apparent if one does not consider complexity of the biochemicals as an asset [13]. In contrast, the production of ethylene from bio-sources is considered. Another pathway suggests the production of synthesis gas, which over Fischer-Tropsch is converted into hydrocarbons.

More effort should be invested into seeking alternatives to the approach of simply replacing the current intermediates. If one considers a change-over process from oil as the main resource to bio-based resources, then one has several options, some of which involve using existing processes, whilst others will be completely new processes. The currently ongoing projects aiming at alternative processes in the EU and the US point towards a chemistry that is based on a few basic chemicals. The report from the US Dept. of Energy [14] can be seen as representative. Here one looks into the feasibility of twelve building block chemicals produced from sugar. These building blocks are 1,4-diacids (succinic, fumaric and malic), 2,5-furan dicarboxylic acid, 3-hydroxy propionic acid, aspartic acid, glucaric acid, glutamic acid, itaconic acid, levulinic acid, 3-hydroxybutyrolactone, glycerol, sorbitol, and xylitol/arabinitol. The key though is how to get the sugar in the first place. Besides the mentioned hydrolysis process, more sophisticated methods are gaining ground by using techniques borrowed from nature. Recently a good number of reviews were published that report on the current status of bio-refineries with an emphasis on biological processing [15, 16, 17, 18]. A success story has recently been reported as a small Dutch company succeeded to enter the market selling its polymers based on 2,5-furan-dicarboxylic-acid (FDCA) to Coca Cola: They will produce the environmentally friendly future Coca Cola bottles [19].

3. Future bio-refineries

The view on bio-refineries is continuously changing as technology and economy open up new opportunities. The governmental energy departments around the world are developing new views on what a bio-refinery could or will constitute. In the EU, the report of the Star-Colibri organisation provides a good picture on how the EU-research and development community looks at the future [20, 21, 22, 23]. In the US the various related departments publish their respective findings and document their ongoing initiatives [24].

Moving towards bio-refineries is certainly a good effort, but economy has not really caught on to enable and support the change. In the current market, the bio-derived products are apart of a few exceptions not yet economically viable, though this could easily change by applying a different pricing on oil.

The future of bio-refining must meet the two objectives, namely (i) material and energy closure on the time scale that is dictated by consumption and (ii) optimized processes being embedded in a large-as-possible system boundary. This implies that a holistic approach where sustainability of the entire value chain needs to be the benchmark. Such a framework will consist of a combination of:

- Foster bio-refining where a palette of chemicals, materials and fuel are produced from biomass (ligno-cellulosics, aquatic biomass, agro-wastes and waste streams).
- Develop chemicals which either substitute or replace existing fossil based chemicals.
- Control economy so that the production of substitutes and/or replacements from biomass become economical viable.
- Optimize the production with respect to material waste and energy.

So it will be necessary to not only change the resources, but also to change the processes aiming at the direct utilization of complex chemical components in contrast to the classical route of breaking-down followed by a synthesis. Also we must develop new materials and new material integration concepts, thus develop whole new production chains. On the operations side, the processes must be optimized to at least the same extent to which we have optimized the petro-based plants. In generic terms one has four major options:

- Digest: the complex molecules are broken down into usable small pieces primarily sugars. From here various options are open.
- Destruct: the complex molecules are broken down into the basic constituents from which through synthesis the products are constructed.
- Separation: Complex molecules are extracted and purified to be used directly as products or are further modified.
- Modification: Easily accessible chemicals are being modified to form desirable products.

4. What is needed?

Primarily investment into the future is needed. The political bodies need to create a positive climate that promotes and maintains a co-operative approach to research and eventually development. On the top level, the future is not in competition but in collaboration, since the world has gone global.

4.1. Research

Activities that focus on ”ecological invisible bio-refining” operating are:

- Cycles that are closed in terms of mass and which solely import energy that is directly or indirectly drawn from the solar energy influx.
- Criteria to measure sustainability considering
  - Site and resource selection based on optimal trade-off between food, fuel and chemical products.
  - Effect on the embedding ecosystems.
  - Handling of waste streams, including all energy streams.
  - Utilization of waste streams.
  - Recycling of nutrients, with ”nutrients” being very broadly defined including fertilizing and supply of trace components.
  - No contamination and no negative long-term effects.
  - Logistics: Transportation vs location of the production vs. distribution.
  - Achieved ”quantum leap”: relative value of the resource and derived product.
  - Sustainability of the resource production (green plant farming, forestry, maritime products such as algae and microbe farming)
  - Efficient multi-faceted conversion appropriately utilizing the (local) resources, such as thermo-chemical, bio-technological of enzymatic treatment.
- Integration with other, non-chemical activities associated with bio-materials mainly aiming at utilizing its waste streams.

Thus research towards an ecological invisible bio-refinery requires exploration of:

- new, alternative chemical pathways aiming at retaining chemical complexity,
- substituting petro-chemical-based products and
- new products.

Succeeding in this research endeavour relies on both experimental approaches as well as computational engineering approaches, the latter requiring the development of new tools that allow for rapid generation and analysis of alternative plants and approaches. Further, comprehensive analysis consisting of a simultaneous environmental and economical analysis is vital.

4.2. Technological challenges of a Bio-refinery

Comprehensive innovation is needed to make the two major routes for bio-refinery processes, biochemical and thermo-chemical conversion, cost effective, sustainable and robust for varying feed stocks and output products. It is necessary to minimize waste and emissions during the conversion processes and to minimize environmental and economic costs.

- Major challenges of *biochemical conversion* of biomass to products are the complexity of unit operations which need to be optimized each for itself and within the entire plant. For a highly efficient biochemical conversion the following issues need to be addressed: efficient pre-treatment for initial extraction of valuable chemicals, subsequent improved hydrolysis processes to improve sugar yield and reduce processing time, more "robust" micro organisms for the conversion of sugars to products and finally optimized novel separation processes to remove products from the fermentation broth. The spectrum of products to be produced biochemically from biomass derived sugars is steadily increasing, however their environmental and economic cost need to be verified. Improving the flexibility of the processes with respect to feedstock utilization and product output is needed. The degree of flexibility is dependent on the market situation. A possible change from production of e.g. an energy carrier (ethanol or butanol) to other marketable products (commodity chemicals or animal feed) and/or their effective co-production, would increase the economic robustness of the refinery.

- Major challenges of *thermo-chemical conversion* of biomass to products, are the high humidity and oxygen content of the biomass raw material, its general recalcitrance towards thermochemical treatments and the intractability of lignin, the main biomass derived residue from ligno-celluloses. Upgrading of the generated condensible fractions includes innovative separation and purification technology, catalytic de-oxygenating and hydrogenating processes. Improved gasification processes, where the ratio of CO/H_2 can be adjusted during operation, such that they are flexible enough to cope with varying feedstocks and respond to varying product demand. Advanced gas handling systems to avoid catalyst deactivation and degradation caused by feedstock components, like condensation of tar, catalyst deactivation by alkalis and chlorine and sensitivity to high water concentrations. Improved pyrolysis through more flexible and robust technologies to efficiently generate pyrolysis oils as feedstock for bio-fuels, chemicals and (performance) materials.

The dominating challenge of an efficient bio-refinery as a whole is the requirement of simultaneous flexibility with respect to feedstock and number of products to ensure economic robustness. Enabling this requires:

- Separation of broth / intermediates / by-products and products will need special attention to reduce energy consumption of the process. Process design needs to be integrated into the search for new products.

- Advanced process integration schemes to enable flexible production, product changeover and process integration.

- Integration of biochemical and thermochemical conversion pathways, to improve material and energy integration needs consideration.

- Process intensification and optimization methods tailored for bio-refineries need to be emphasized.

Considering the number of highly complex and interrelated research issues presented makes it apparent that there is a comprehensive need for basic and applied scientific performed in universities, research institutes and industries to improve the environmental and economical efficiency of a green refinery.

5. Discussion

The challenges in a transition from a fossil-carbon based production to a sustainable renewable-carbon based production are prominent. The dominant challenge is a need for a new orientation of the producing

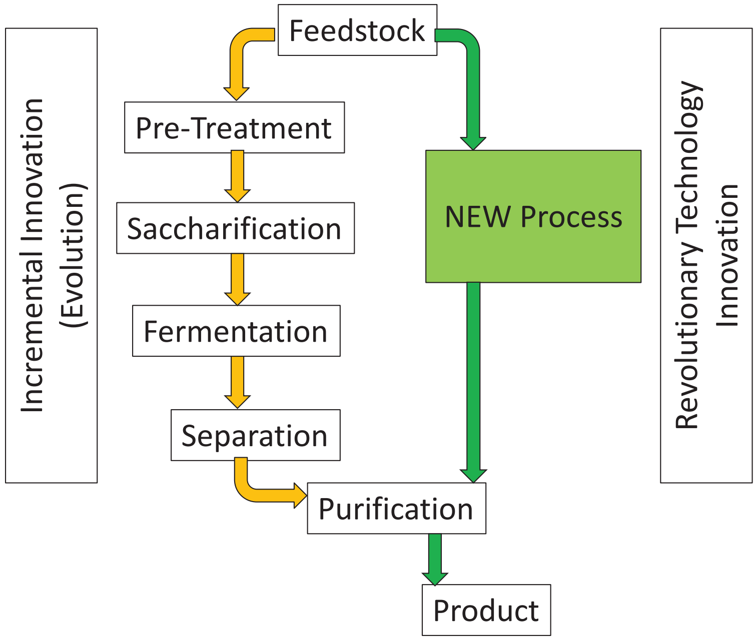

Fig. 1. Incremental versus revolutionary Innovation

industry, directed towards society, which is built on sustainability and utilization of renewable sources. Based on sustainable and available resources, new structures are generated which, provided the political incentives are given and maintained, will enable a transition from fossil-based to biomass-based, products. Thus a controlled transition based on *foresight* could be initiated which considers the most prevailing input factors.

Without political incentives promoting the needed change, today's existing economical framework driving our industries is also the stumbling block inhibiting a change of direction. The return on investments for new processing capacity of a well known process is generally considerably higher than the introduction of an entirely new process. An incremental innovation ensures the short to medium term economical viability of an existing production through e.g. an increase in efficiency. A revolutionary innovation, e.g. combining a novel process with a new renewable feedstock, for a replacement/substitution product, even with only slightly differing properties, requires venture capital willing to take a considerable risk.

A balanced combination of incremental innovation and revolutionary innovation (see Figure 1) for the production of energy, chemicals, materials, feed and food in green refineries might be a suitable pathway of transition. Alternative enabling scenarios are the following:

- Utilization of the known bio- and thermochemical conversion pathways for the production of existing products based on new feedstocks or products to substitute the original products.
- Intensified research on the modification of plants to produce products or intermediates with considerable higher yield
- Comprehensive research to replace or substitute known products and materials (e.g.: polyethylene is substituted by polylactic acid).
- Development of multi-product production process to enhanced raw material utilization.

These scenarios can be implemented by systematic utilization of the modern chemistry, which allows for digestion (complex molecules are converted into usable products), destruction (breakdown to small compounds and their recombination), separation (removal of valuable chemicals) and reconfiguration (modify complex structures). However, utilization of these basic conversion steps for the production of biomass based products needs to be integrated or at least combined with today's technology to enable a transition. The needed transitions are facilitated through replacement of the mined carbon by biomass and production of the same intermediates, thus feeding into existing processes and produce mostly the same products. This requires new process developments for the conversion of the available biomass to known intermediates for further processing, thereby limiting the risk for the pretreatment process. The remaining process requires limited changes, thus represents a confined economical risk. Parallel to the co-feed strategy, new conversion paths are needed for the production of important products. This may not necessarily mean new chemistry, but to use more bio-based products, such as the use of organic fibres in composite products or the like.

Further, the integration must aim at utilizing waste streams from other industrial activities, thereby generating a symbiotic effect that makes both industrial branches economically more viable. For example, an increased use of wood for construction purposes will produce more wood waste, which can be utilized in bio-refining activities. A major challenge is to find a good balance between transportation and processing. The low costs of fuel have promoted a centralized approach to processing, a philosophy, which more than likely will have to be reverted as bio-material is bulky, heavy and refining processes require large quantities of materials. It is thus advisable to focus on approaches that have a good chance to be viably realizable in relatively small scales.

6. Conclusion

Assuming that humanity indeed does switch away from mined carbon resources and increasingly relies on carbon being in the biosphere cycle, we believe that the current policy of replacing fuel and feedstock for the classical chemical production is not rich enough to address the problem appropriately. This approach may result in some improvement but it certainly is too small of a step to make the overall system, these days being the world as a whole, a sustainable system. We believe that any lasting development will have to implement a policy that follows this paradigm:

Sustainability can only be achieved if the human activity remains within the biosphere we are part of and when the time scale of usage and regeneration match.

And that we adjust the goals for the operations to:

Complex molecules should be retained and built on in contrast to breaking things down to what we currently consider base stock.

Whilst we have the technology to synthesize from primary feedstock and purification, the retaining operation requires only separation. Overall, this will be more energy efficient and resulting in less side effects, whilst possibly requiring more sophisticated separation processes. Certainly the energy requirements are significantly lower and thus secondarily also contribute to improved sustainability.

Strong research efforts are necessary. History tells us that many ideas and processes will never be realized for either economical or technical reasons. Success is measured on the realised process and conditions that effect the product/measures. Further, measures themselves change with time, all of which can usually not be predicted reliably. Finding likely winners is certainly not an easy task. Thus many things must be tried and tested; and success must consequently carry the effective costs including the ones that were failures.

Strong integration with other sectors must be sought and small-scale operations that integrate with distributed bio-related production processes must be given priority.

The different transition stages cannot be pursued all at once. It will be necessary to do things in stages. The first stage should aim at substituting fuel and replace the intermediates entering the chemical production with intermediates produced from bio-sources. At the same time alternative productions must be faced in essentially taking over all the petro-based production processes. Critical in the whole process is time: development time for known processes is at least 10 years, whilst new processes require in the order of 20 years to be established in industries. The change into processes where we have little to no idea on what they are is more in the framework of 50 to 100 years. It seems that it is high time that we start to put our head onto the tasks and explore alternatives.

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